

# 12.42% Monolithic 25.42 cm<sup>2</sup> Flexible Organic Solar Cells Enabled by an Amorphous ITO-Modified Metal Grid Electrode

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Printed metal nanogrid electrode exhibits superior characteristics for use in flexible organic solar cells (OSCs). However, the high surface roughness and inhomogeneity between grid and blank region is adverse for performance improvement. In this work, a thin amorphous indium tin oxide (ITO) film ( $\alpha$ -ITO) is introduced to fill the blank and to improve the charge transporting. The introduction of  $\alpha$ -ITO significantly improves the comprehensive properties of metal grid electrode, which exhibits excellent bending resistance and long-term stability under double 85 condition (under 85 °C and 85% relative humidity) for 200 h. Both experimental and simulation results reveal  $\alpha$ -ITO with a sheet resistance of 20 000  $\Omega \square^{-1}$  is sufficient to improve the charge transporting within the adjacent grids, leading to a remarkable efficiency of 16.54% for 1 cm<sup>2</sup> flexible devices. With area increased to 4.00, 9.00, and 25.42 cm<sup>2</sup>, the devices still display a performance of 16.22%, 14.69%, and 12.42%, respectively, showing less efficiency loss during upscaling. And the 25.42 cm<sup>2</sup> monolithic flexible device exhibits a certificated efficiency of 12.03%. Moreover, the device shows significantly improved air stability relative to conventional high-conductive poly(3,4-ethylenedioxythiophene):polystyrene sulfonate-modified device. All these make the  $\alpha$ -ITO-modified Ag/Cu electrode promise to achieve high-efficient and long-term stable large-area flexible OSCs.

mechanical flexibility, lightweight, compatibility with roll-to-roll manufacturing processes, and capability for building integrated photovoltaic applications.<sup>[1–8]</sup> The highest power conversion efficiency (PCE) of small-area single junction and tandem flexible OSCs has reached above 16% now.<sup>[2,9–11]</sup> However, the PCE of 1 cm<sup>2</sup> flexible cells was only 14.29%,<sup>[12]</sup> lagging those small-area devices. For the practical application of flexible OSCs, the efficiency improvement of large-area flexible devices is essential.<sup>[13–15]</sup>

With the scale-up of flexible OSCs, the electrical loss from large sheet resistance of the flexible transparent electrode (FTE) causes a dramatic performance drop. FTEs for the flexible OSCs should have an overall property of low sheet resistance, high transmittance, and excellent mechanical stability. Till now, several FTEs, including conducting polymer,<sup>[9,10,16–19]</sup> carbon nanotube,<sup>[20,21]</sup> graphene,<sup>[22–24]</sup> thin metal,<sup>[25–27]</sup> metallic nanowires,<sup>[2,28–30]</sup> metal grid<sup>[14,31,32]</sup> have been successively

applied in flexible OSCs. Among them, the metal grid electrode has a lowest sheet resistance ( $R_s$ ) as low as 1  $\Omega \square^{-1}$ .<sup>[32–36]</sup> The metal grid electrodes can be fabricated through thermal evaporation,<sup>[37,38]</sup> inkjet-printing,<sup>[39–41]</sup> and imprinting.<sup>[42–44]</sup> The

## 1. Introduction

Flexible organic solar cells (OSCs) have attracted enormous consideration due to their outstanding traits, including

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